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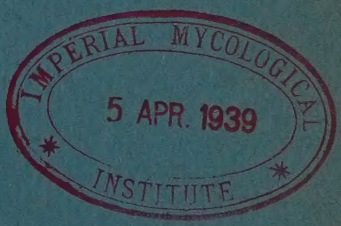

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MATHEMATICS

A NEW IMPROVEMENT OF THE METHOD OF ESTIMATION OF TRIGONOMETRICAL SUMS WITH PRIMES

By I. M. VINOGRADOW, Member of the Academy

By slight modification of my method developed in 1937 and in particular by addition of some elements of the method due to Viggo Brun, it is possible to enlarge the region of application of my method. I give here some results in this direction. The complete proofs will appear the next future.

1. Let n be a constant integer $\geqslant 1$, ε — an arbitrary small positive constant < 1, h — an arbitrary large constant > 1, N — an integer > 2, p runs over primes,

$$S = \sum_{p \leqslant N} e^{2\pi i \alpha p^n},$$

$$\alpha = \frac{a}{q} + \frac{\theta}{q\tau}, \quad (a, q) = 1, \quad (\log N)^{\varepsilon} < q \leqslant (\log N)^h,$$
$$\tau = N^n (\log N)^{-h}, \quad -1 \leqslant \theta \leqslant 1.$$

Then we have

$$|S| < c \frac{N}{\log N} q^{-\delta},$$

where δ is positive depending only on n and c depends only on n, ε and h.

Particular Case. Let ε and η be arbitrary small positive constants < 1, h — an arbitrary large constant > 1, N — an integer > 2, p runs over primes,

$$S = \sum_{p \leqslant N} e^{2\pi i \alpha p},$$

$$\alpha = \frac{a}{q} + \frac{\theta}{q\tau}, \quad (a, q) = 1, \quad (\log N)^{\varepsilon} < q \leqslant (\log N)^h,$$
$$\tau = N (\log N)^{-h}, \quad -1 \leqslant \theta \leqslant 1.$$

Then we have

$$|S| < c \frac{N}{\log N} q^{\eta - \frac{1}{2}},$$

where c depends only on ε, η and h.

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MATHEMATICS

ON THE LIMITING THEOREMS FOR SUMS OF INDEPENDENT VARIABLES

By B. GNEDENKO

(Communicated by S. N. Bernstein, Member of the Academy, 27. XI. 1938)

As in the foregoing note (1), we shall consider a sequence of series of random variables

$$x_{n,1};\ x_{n,2};\ \ldots;\ x_{n,k_n},\quad (n=1,2,3,\ldots) \tag{1}$$

independent within each series and negligible in the limit, i. e. such that uniformity with respect to k $(1 \leqslant k \leqslant k_n)$

$$\lim_{n\to\infty} P\{|x_{n,k}| > \varepsilon\} = 0$$

for any $\varepsilon > 0$. In the present note we give new necessary and sufficient conditions for the convergence of the distribution laws of sums

$$s_n = x_{n,1} + x_{n,2} + \ldots + x_{n,k_n} \tag{2}$$

to some limiting law. We now establish necessary and sufficient conditions which, different from those given in the foregoing note, involve directly the distribution functions $F_{n,k}(x)$ of the $x_{n,k}$. The detailed proofs of the following results will be published in the «Bulletin de l'Académie des Sciences de l'URSS».

The Fundamental Theorem. *In order that the sequence of distribution laws of the sums* (2) *should converge to some limiting distribution law* $\Phi(x)$, *it is necessary and sufficient that there should exist non-decreasing functions* $M(x)$ *and* $N(x)$ *defined on* $(-\infty, -0)$ *and* $(+0, +\infty)$ *respectively, and constants* a, τ, $\gamma(\tau)$, *satisfying the following conditions*:

1) $-\tau$ *is a point of continuity of* $M(x)$, $+\tau$ *is a point of continuity of* $N(x)$;

2) $M(-\infty) = N(+\infty) = 0$

$$\int_{-\tau}^{-0} x^2 dM(x) + \int_{+0}^{+\tau} x^2 dN(x) < +\infty;$$

3) $$\sum_{k=1}^{k_n} F_{n,k}(x) \xrightarrow[n\to\infty]{} M(x) \quad \text{for } x < 0,$$

$$\sum_{k=1}^{k_n} (F_{n,k}(x) - 1) \xrightarrow[n\to\infty]{} N(x) \quad \text{for } x > 0$$

at all points of continuity of the functions $M(x)$ *and* $N(x)$;

$$4)\quad \sum_{k=1}^{k_n} \int_{|x|<\tau} x\,dF_{n,k}(x) \xrightarrow[n\to\infty]{} \gamma(\tau)$$

and

$$5)\quad \lim_{\varepsilon\to 0}\lim_{n\to\infty} \sum_{k=1}^{k_n} \left\{ \int_{|x|<\varepsilon} x^2\,dF_{n,k}(x) - \left(\int_{|x|<\varepsilon} x\,dF_{n,k}(x) \right)^2 \right\} = a^2.$$

Observe that in the case when the conditions of the theorem are satisfied for some τ, they are automatically satisfied for any other τ, for which $-\tau$ and $+\tau$ are points of continuity of the functions $M(x)$ and $N(x)$ respectively, and the corresponding value of the constant $\gamma(\tau)$.

When the conditions of the theorem are satisfied, the limiting distribution function itself is determined by its characteristic function $\varphi(t)$ which may be found by P. Lévy's formula:

$$\lg \varphi(t) = i\gamma(\tau)\,t - \frac{a^2}{2}t^2 + \int_{-\infty}^{-\tau} (e^{itu}-1)\,dM(u) + \int_{+\tau}^{+\infty} (e^{itu}-1)\,dN(u) +$$

$$+ \int_{-\tau}^{-0} (e^{itu}-1-itu)\,dM(u) + \int_{+0}^{+\tau} (e^{itu}-1-itu)\,dN(u). \qquad (3)$$

Formula (3) shows that the limiting distribution law $\Phi(x)$ necessarily belongs to the class of the so-called infinitely divisible distribution laws. This fact was established earlier by A. Khintchine (2).

From the formulated fundamental theorem we can easily deduce the condition for the convergence of the distribution laws of the sums (2) to different special limiting laws. We state here several theorems of this type.

Theorem 1. *In order that the distribution laws of the sums* (2) *should converge to Gauss' law*

$$\Phi(x) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x} e^{-\frac{z^2}{2}}\,dz$$

and that the variables $x_{n,k}$ *should be negligible in the limit, it is necessary and sufficient that the following conditions should be satisfied for every* $\varepsilon > 0$:

$$1)\quad \sum_{k=1}^{k_n} \int_{|x|\geqslant\varepsilon} dF_{n,k}(x) \xrightarrow[n\to\infty]{} 0;$$

$$2)\quad \sum_{k=1}^{k_n} \int_{|x|<\varepsilon} x\,dF_{n,k}(x) \xrightarrow[n\to\infty]{} 0;$$

$$3)\quad \sum_{k=1}^{k_n} \left\{ \int_{|x|<\varepsilon} x^2\,dF_{n,k}(x) - \left(\int_{|x|<\varepsilon} x\,dF_{n,k}(x) \right)^2 \right\} \xrightarrow[n\to\infty]{} 1.$$

Proof. Since for the law $\Phi(x)$ we must put in the formula (3) $M(u) \equiv N(-u) \equiv 0$, $\gamma(\tau) = 0$, $a = 1$, the fundamental theorem gives the conditions of Theorem 1 without any calculations. Theorem 1 is substantially equivalent to a theorem of W. Feller (3).

Theorem 2. *In order that the distribution laws of the sums* (2) *of negligible in the limit variables should converge for* $n \to \infty$ *to Poisson's law*

$$P(x) = \sum_{0 \leq k < x} \frac{e^{-\lambda} \cdot \lambda^k}{k!},$$

it is necessary and sufficient that the following conditions should be satisfied for every $\varepsilon > 0$:

$$1) \quad \sum_{k=1}^{k_n} \int_{R_\varepsilon} dF_{n,k}(x) \xrightarrow[n \to \infty]{} 0;$$

$$2) \quad \sum_{k=1}^{k_n} \int_{|x-1| < \varepsilon} dF_{n,k}(x) \xrightarrow[n \to \infty]{} \lambda$$

$$3) \quad \sum_{k=1}^{k_n} \int_{|x| < \varepsilon} x dF_{n,k}(x) \xrightarrow[n \to \infty]{} 0;$$

$$4) \quad \sum_{k=1}^{k_n} \left\{ \int_{|x| < \varepsilon} x^2 dF_{n,k}(x) - \left(\int_{|x| < \varepsilon} x dF_{n,k}(x) \right)^2 \right\} \xrightarrow[n \to \infty]{} 0,$$

where R_ε *denotes the set* $|x| \geqslant \varepsilon$, $|x-1| \geqslant \varepsilon$.

Proof. For the law $P(x)$ in P. Lévy's formula we have to put $a = \gamma(\tau) = 0$, $M(u) \equiv 0$, $N(u) = 0$ for $u < 1$, $N(u) = 1$ for $u > 1$. Then it becomes clear that Theorem 2 is an obvious consequence of the fundamental theorem.

Theorem 2 was found simultaneously with and independently from me by Marzinkiewicz (4).

Theorem 3. *In order that for any* $\varepsilon > 0$ *we should have*

$$P\{ |x_{n,1} + x_{n,2} + \ldots + x_{n,k_n}| \geqslant \varepsilon \} \xrightarrow[n \to \infty]{} 0$$

and that the independent variables $x_{n,k}\ (1 \leqslant k \leqslant k_n)$ *should be negligible in the limit, it is necessary and sufficient that the following conditions should be satisfied for every* $\varepsilon > 0$:

$$1) \quad \sum_{k=1}^{k_n} \int_{|x| > \varepsilon} dF_{n,k}(x) \xrightarrow[n \to \infty]{} 0;$$

$$2) \quad \sum_{k=1}^{k_n} \int_{|x| < \varepsilon} x dF_{n,k}(x) \xrightarrow[n \to \infty]{} 0;$$

$$3) \quad \sum_{k=1}^{k_n} \left\{ \int_{|x| < \varepsilon} x^2 dF_{n,k}(x) - \left(\int_{|x| < \varepsilon} x dF_{n,k}(x) \right)^2 \right\} \xrightarrow[n \to \infty]{} 0.$$

Proof. Theorem 3 follows directy from the fundamental theorem for

$$\Phi(x) = \begin{cases} 0 \text{ for } x < 0 \\ 1 \text{ for } x > 0. \end{cases}$$

Theorem 3 is substantially equivalent to the necessary and sufficient conditions for the law of great numbers given by A. Kolmogoroff (5) and W. Feller (6).

Theorem 4. *In order that for non-negative random variables* $x_{n,k}$, *independent within each series, we should have for any* $\varepsilon > 0$

$$P\{|x_{n,1}+x_{n,2}+\ldots+x_{n,k_n}-1|>\varepsilon\} \xrightarrow[n\to\infty]{} 0$$

and that the $x_{n,k}$ *should be negligible in the limit, it is necessary and sufficient that the following conditions should be satisfied for every* $\varepsilon > 0$:

$$1)\quad \sum_{k=1}^{k_n} \int_{\varepsilon}^{\infty} dF_{n,k}(x) \xrightarrow[n\to\infty]{} 0;$$

$$2)\quad \sum_{k=1}^{k_n} \int_{0}^{\varepsilon} x\,dF_{n,k}(x) \xrightarrow[n\to\infty]{} 1.$$

Proof. Theorem 4 follows from the fundamental theorem, if for the limiting law of the distribution laws of the sums (2) we take

$$\Phi(x) = \begin{cases} 0 \text{ for } x < 1 \\ 1 \text{ for } x > 1. \end{cases}$$

This theorem is equivalent to the necessary and sufficient conditions for the relative stability of the sums found by A. Bobrov (7).

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BIOPHYSICS

CRITICAL FREQUENCY OF FLICKER AND INDIRECT STIMULI

By S. V. KRAVKOV

(Communicated by L. A. Orbeli, Member of the Academy, 4. XII. 1938)

The determination of the critical frequency of flicker, that is such a frequency which gives the impression of integral non-flickering light, is applied in the study of physiological optics as one of the methods characterizing the condition of the sight organ depending upon adaptation [(1), (2) and others], fatigue (3), pathological conditions (4), drug influence (5). The substratum of flicker frequency remains, however, far from being adequately elucidated.

The method of indirect stimuli, which we have lately adapted for the study of various aspects of vision, permitted us of establishing certain regularity in the changes of the critical frequency of flicker. It seems to us that the data obtained may throw light upon the general problem of the nature of critical frequency of flicker.

The aim of our experiments was to determine the critical frequency of flicker for foveal vision in the course of a 60-minute dark adaptation. Monochromatic rays of different wave lengths served as light stimuli. The indirect stimulus began to act upon the subject about 30 minutes after the beginning of adaptation; it lasted 10 minutes. In experiments bearing upon the problem of our article, we used as indirect stimuli in some cases oleum Bergamotae, in others—sound. When odour was to act as indirect stimulus, an open bottle which contained cotton soaked in oleum Bergamotae was placed close to the nostrils of the subject. The odour was sufficiently strong. In the test where sound was used, the latter was derived from a lamp-generator which had a frequency of about 800 Hertz and a loudness level of about 85 decibels. The sound was transmitted to the ears of the subject with the help of telephone receivers.

As a rule, the definitions of the critical frequency of flicker were recorded every 7 or 8 minutes during the first half hour of the dark adaptation; during the time of the indirect stimulus the definitions were recorded more often—every two minutes. In all, during the action of the indirect stimulus upon the subject three definitions of the critical frequency of flicker were recorded. After the indirect stimulus had ceased to act, the experiment went on for some 20 or 25 minutes, during which two more definitions of the critical frequency of flicker were effected. Thus, by comparing the level of the critical frequency of flicker, determined under the influence of the indirect stimulus, with that previous to and following the action of the indirect stimulus, it proved possible to judge about the trend of the change in the critical frequency of flicker as influenced by a given indirect stimulus.

These experiments, carried out on seven subjects, gave the following results, which proved to be in full agreement: under the influence of the odour of oleum Bergamotae, as well as under the influence of sound, the critical frequency of flicker increases for the red colour with wave lengths of 160, 630 and 650 mμ, and with a brightness corresponding to the initial critical frequency of 14—18 per/sec.; whereas for the green with wave length of 520 mμ of the same intensity, the critical frequency of flicker under the influence of the same indirect stimuli decreases.

However, our previous experiments (1936) on the change produced in the colour sensitivity of the eye under the influence of sound stimuli have shown (6) that the sensitivity of the eye to the green colour rises under the influence of sound, whereas its sensitivity to the orange-red decreases. Thus, the comparison of these data with those exposed in this article

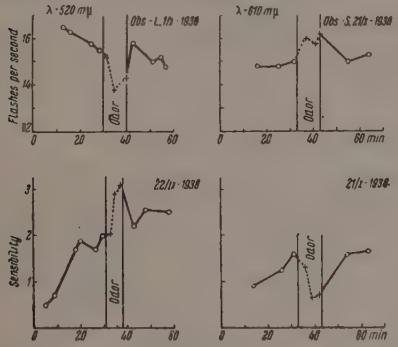

Fig. 1. The effect of one and the same stimulus upon the frequency of flicker and upon the colour sensitivity. The lower curves show a change in the sensitivity; the upper—a change in critical frequency of flicker.

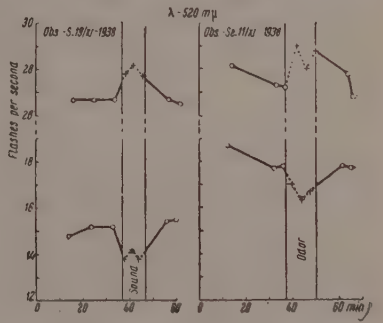

Fig. 2. The action of one and the same indirect stimulus upon the critical frequency of flicker of light of different intensity.

makes us believe that an increase in the critical frequency of flicker under the influence of indirect stimuli must not necessarily point to an increase of the sensitivity of the eye of the subject.

We also performed special tests, which involved not only determination of the critical frequency of flicker during the action of an indirect stimulus (odour or sound), but the colour sensitivity as well. The latter was determined by the method of the extinction of the colour ray, up to the disappearance of any noticeable chromaticity.

The typical results obtained are illustrated by the records given in Fig. 1.

It may be seen that the indirect stimuli may heighten the sensitivity of the eye and simultaneously diminish the critical frequency of flicker, and vice versa. However, with the increase of sensitivity weak stimuli behave as objectively stronger ones, while the objective heightening of the light stimulus according to Ferri-Porter's law leads to an increase in the frequency of flicker. Therefore, with regard to the critical frequency of flicker, the increase in the sensitivity of the eye under the influence of an indirect stimulus should not be identified with the increase in the objective intensity of the stimulus. Whereas the sensitivity of the eye under the effect of an objective intensification of the stimulus during the period of darkening remains reduced (because of the considerable expenditure of light sensitive substances caused by an intensified light

stimulus), in the case of its heightening under the influence of one or another indirect stimulus, the sensitivity proves enhanced not only during the period of the light stimulation, but also during those moments of darkening, when the excitation caused by the light stimulus extinguishes. Owing to such a heightened sensitivity of the nervous apparatus, the relative difference between the levels of excitation at the period of illumination and excitation, which is also present during darkening, may diminish. One may easily conclude that the latter may result in a decrease of the critical frequency of flicker.

Such is the probable process of action of indirect stimuli upon the critical frequency of flicker, insofar as the indirect stimuli change the sensitivity (excitability) of our vision.

However, our tests on the effect of indirect stimuli upon the contrast-sensitivity of the eye and the irradiation, published a few years ago ([7]), lead us to the conclusion that indirect stimuli may change not only the sensitivity of the eye, but its excitation (or excitatory state) as well. It was found that an increase in the sensitivity caused by indirect stimuli is distributed over the directly excited zone the more unevenly, the more uneven is the direct excitation.

From the above it should be assumed that one and the same indirect stimulus may affect the critical frequency of flicker in a different manner, depending upon the intensity of the flickering light. If the light is bright and if the unevenness of the excitation during the period of illumination and darkening is considerable, then this unevenness may be intensified by the indirect stimulus. This will result in an increase of the critical frequency of flicker. If, on the other hand, the flickering light is dim, then the unevenness of excitation of the vision, created by it, will be insignificant, and thus the excitation due to the influence of indirect stimulus may still diminish the unevenness. As a result, a reduced critical frequency of flicker is obtained. Special tests (with green light) performed by us have corroborated the above assumption. It actually appeared that one and the same indirect stimulus (odour or sound) may increase the critical frequency of flicker of a bright light and diminish that of a weak one.

The records of this experiment are given in Fig. 2.

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BIOPHYSICS

ON SOME CORRELATIONS OF DIFFERENT RECEPTORS IN OUR COLOUR VISION

By S. V. KRAVKOV

(Communicated by L. A. Orbeli, Member of the Academy, 4. XII. 1938)

The experiments we have performed with the changes in the colour sensitivity of the eye under the influence of acoustic stimuli have shown [1] that the sensitivity to the green-blue rays of the spectrum is enhanced when acted upon by sounds, while the response to orange-red is, on the contrary, reduced. When applying as direct stimuli different rays of the spectrum, we found the effect of indirect sound excitation (both positive and negative) to decrease with the approach of the direct stimulus to the red or violet ends of the spectrum.

Yakovlev's [2] experiments have established that the visual field for the orange-red stimulus is subject to noticeable restriction under the influence of an indirect sound excitation, while for an extreme red stimulus it remains unchanged under the same conditions.

Finally, the experiments performed by Dobryakova[3] in our laboratory with the effect of taste stimuli (sugar) on the critical frequency of flicker of various monochromatic rays, made it evident too that the eye shows no reaction whatever to indirect taste excitation when acted upon with extreme red stimulus, whereas the critical frequency of flicker for the orange-red stimulus undergoes a definite change under similar conditions.

In order to give interpretation to the above facts, we had recourse to the following hypothesis: the red excitation reacts to the applied indirect stimuli only when the green sensitive receptor of the eye is to a sufficient degree affected by it. If, on the other hand, the green sensitive receptor fails (altogether or almost) to be touched by the given red stimulus, then there is no reaction to the indirect stimulation. In other words, we come to assume that the red sensitive receptor of the eye itself displays no direct reaction to indirect stimulation, but is able to undergo changes only in the case of simultaneous functioning of the green sensitive receptor which can be directly transformed by the indirect stimuli.

Our experiments, as well as those performed by Yakovlev, have established a decrease in the colour senstitivity to the orange-red stimulus under the influence of indirect sound stimuli. Dobryakova's experiments have confirmed the increase in the critical frequency of flicker for the orange-red rays under the influence of indirect taste stimuli. In our special experiments[4], in which we made parallel measurements both of the critical frequency of flicker and the colour sensitivity under the influence of analogous indirect

stimuli, we have shown that an increase in the critical frequency of flicker when affected by indirect stimuli takes place parallel to a decrease in colour sensitivity and vice versa.

We can thus somewhat specify the above formulation of our hypothesis by stating that the excitability (sensitivity) of the red sensitive receptor of our vision decreases under the influence of indirect stimuli (applied in our experiments) only in the case when there is a heightening in the excitability (sensitivity) of the green sensitive receptor.

We have also performed special experiments with the effect of odour applied as an indirect stimulus (the odour of oleum Bergamotae) upon the critical frequency of flicker of the orange-red light with a wave length of 600 mμ and of the red light with a wave length of 690 mμ.

The critical frequency of flicker for foveal vision was determined in the course of dark adaptation. The experiments performed upon two subjects showed in full agreement with each other that the critical frequency of flicker with a wave length of 600 mμ increased under the influence of the odour, whereas the critical frequency of flicker for the red light with a wave length of 690 mμ under similar conditions gave evidence of no change whatsoever.

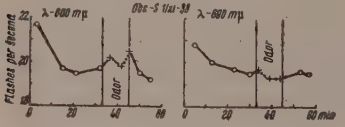

Fig. 1. Effect of one and the same indirect stimulus on the critical frequency of flicker of the red (600 mμ) and extreme red (690 mμ) light.

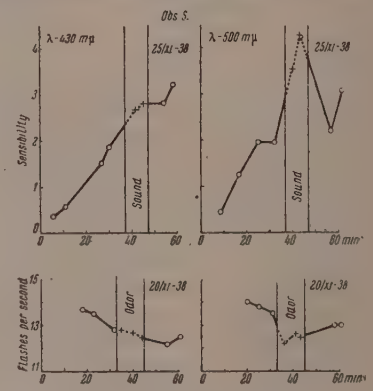

Fig. 2. Effect of one and the same indirect stimulus on the colour sensitivity (upper curves) and upon the critical frequency of flicker (lower curves) of green (500 mμ) and violet (430 mμ) light.

Fig. 1 shows the data of the tested C. as obtained in one set of the experiment. The number of periods of light corresponding to the critical frequency of flicker are plotted along the ordinate, along the abscissa—dark adaptation in minutes.

After it had been established in all above experiments that a change in the reaction of the red sensitive receptor of the eye occurs under the influence of indirect stimuli in that part of the spectrum, which touches upon the green sensitive receptor, another question arose.

Namely: it appeared of interest to find out whether the variability of the reaction of the eye to the short waves of the spectrum, in case indirect stimuli are applied, is also determined by the green sensitive receptor of the eye.

In order to answer this question, we performed experiments with the effect of sound used as an indirect stimulus in some cases and odour in others upon the colour sensitivity and the critical frequency of flicker of foveal vision. Both colour sensitivity and critical frequency of flicker were determined during dark adaptation. Monochromatic rays with wave lengths

of 500 mμ (blue-green) and of 430 mμ (violet) were applied as direct light stimuli. While the former affect to a considerable degree the green sensitive receptor of the eye, the violet rays with a wave length of 430 mμ do not act upon it at all.

The results of the experiments prove to be concordant; they have shown that the reaction of the eye to the short waves which remain untouched by the green sensitive receptor of the eye is not in the least changed under the influence of the indirect stimuli applied, whereas with regard to the blue-green rays, under the influence of the same indirect stimuli, the colour sensitivity of the eye, as well as the critical frequency of flicker, underwent sharp changes. The data of Fig. 2 illustrate the above-said.

In conclusion, we feel free to assume that the green sensitive receptor of the eye in normal trichromatic vision occupies a special position, in the sense that it is capable of reacting to various indirect stimuli. The decrease of the excitability of the red sensitive receptor under the influence of indirect stimuli should be regarded as a result of the antagonistic action on the part of the green sensitive receptor of the eye, which, when affected by the very same indirect stimuli, experiences enhanced excitability. The blue sensitive receptor of the eye seems not to be directly touched by indirect stimuli either.

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BIOPHYSICS

SOME REGULARITIES UNDERLYING THE EFFECT OF INDIRECT STIMULI (NEBENREIZ) UPON THE DISCRIMINATION SENSITIVITY OF THE EYE

By N. T. FEDOROV

(Communicated by S. I. Wawilow, Member of the Academy, 6. XII. 1938)

The effect of indirect stimuli upon the visual function was studied mainly in the Soviet Union. Most valuable information on this subject has been brought forward by P. P. Lasarev, L. A. Orbeli, B. M. Teplov, A. V. Lebedinski, S. V. Kravkov, K. Cr. Kekcheev, Dionesov, a. o. These studies are chiefly dealing with the effect of indirect stimuli on the threshold sensitivity of the eye. In this connexion great interest should be attached to the phenomenon studied by Teplov and independently by Kekcheev, namely, the inversion of the effect of indirect stimulus with variation of its intensity.

The purpose of the present paper is to subject to a quantitative treatment the principal data available as to the effect of indirect stimuli on the discrimination sensitivity of the eye.

An analysis of these experimental data has shown that all the regularities actually observed can be easily explained as a mere consequence of a general rule which runs as follows: the change in the value of the threshold stimulation upon exposure to an indirect stimulus does not depend on the intensity of the direct stimulus, being determined only by the sensitivity of the brain centres and the intensity of the indirect stimulus.

It will be noted that the stimulus is measured in energy (erg, joule, calorie, kg-m), or proportional units (e. g. in lumena, etc.), while the stimulation is understood by us as the physico-chemical change called forth by the stimulus in the endings of the optic nerve, or, following Adrian, any change of the medium surrounding the excitable tissue which when sufficiently intense can bring the tissue into an excitation state. This stimulation induces in its turn a certain excitation in the optic nerve which, being propagated along the nerve up to the brain, produces a definite sensation. The amount of substance decomposed by the light in the retina, the amount of decomposition products which stimulate the nerve endings, obviously depend on the intensity of the light acting upon the eye, which can be in a general form expressed as follows:

$$x = f(I),$$

whence follows:

$$I = \varphi(x). \qquad (1)$$

Let I be increased by ΔI which causes a hardly perceptible difference in brightness. The following alternatives may be arbitrarily suggested: a) to obtain a scarcely noticeable brightness increment, it is necessary that the increase in the amount of the substance decomposed by light be equal to a certain constant value which depends on the sensitivity of the centres (Cobb's hypothesis), or b) the ratio $\frac{\Delta x}{x}$ should be equal to a certain constant (Lasarev's hypothesis). Taking into account that $(\Delta x)^2$ and $\left(\frac{\Delta x}{x}\right)^2$ are inferior to the threshold value, one obtains from equation (1) by expressing $\varphi(x + \Delta x)$ in terms of infinite series with increasing powers Δx:

$$\Delta I = \varphi'(x)\,\Delta x \text{ and } \frac{\Delta I}{I} = \frac{\varphi'(x)}{\varphi(x)}\,\Delta x = \psi(x)\,\Delta x. \qquad (2)$$

According to the firt hypothesis $\Delta x = \text{const}$. Equation (2) may be presented in the following form:

$$\frac{\Delta I}{I} = \frac{\Delta x}{x}\,x\psi(x) = \frac{\Delta x}{x}\,\psi_1(x), \qquad (3)$$

whence it may be readily seen that with the particular constant I both alternatives lead to the conclusion that Δx for the particular I should be constant, its magnitude depending on the sensitivity of the brain. If the latter is impaired by the indirect stimulus, Δx should be increased, and we may take that in this case

$$\frac{\Delta' I}{I} = (\Delta x + \Delta_1 x)\,\psi(x) = \Delta x\left(1 + \frac{\Delta_1 x}{\Delta x}\right)\psi(x) = \Delta x\,(1+\alpha)\,\psi(x).^* \qquad (4)$$

From formulae (2) and (4) a number of conclusions are derived which can be experimentally checked. By introducing the photometric sensitivity of the eye E as a reciprocal of $\frac{\Delta I}{I}$, we obtain:

$$E_1 = \frac{1}{\Delta x\,\psi(x)} \text{ and } E_2 = \frac{1}{\Delta x\,(1+\alpha)\,\psi(x)}.$$

By dividing E_2 by E_1 and substituting $\frac{1}{E_1\psi(x)}$ for Δx, we have:

$$\frac{E_2}{E_1} = \frac{1}{1+\alpha} = \frac{1}{1+\frac{\Delta_1 x}{\Delta x}} = \frac{1}{1 + E_1\Delta_1 x\,\psi(x)}$$

or

$$E_2 = \frac{E_1}{1 + E_1\Delta_1 x\,\psi(x)}. \qquad (5)$$

Let us consider the following three cases.

I. Let us assume that the indirect stimulus is constant and, consequently, $\Delta' x$ and the intensity of the direct stimulus I, and, consequently, $\psi(x)$ are constant too. Thus

$$E_2 = \frac{E_1}{1 + \alpha E_1}. \qquad (6)$$

In order to check this formula, use was made of S. V. Kravkov's data concerning the influence of sound or illumination of the other eye

* In the case of the increase of brain sensitivity, $\Delta_1 x$ will, obviously, be taken with minus.

on the photometric sensitivity as dependent on the initial photometric sensitivity. In the graph of Fig. 1 Kravkov's data are plotted illustrating the influence of sound; the upper curve refers to the intensity (in relative units) of the direct stimulus $I=3$, the lower one to that of the direct stimulus $I=100$. The squares and triangles stand for experimental data, solid lines for those computed from the formulae:

$$\frac{E_2}{E_1}=\frac{1}{1+0.0043E_1} \qquad \text{(for } I=3\text{)}$$

and

$$\frac{E_2}{E_1}=\frac{1}{1+0.0123E_1} \qquad \text{(for } I=100\text{)}.$$

By illuminating the other eye we obtain in a quite similar way:

$$\frac{E_2}{E_1}=\frac{1}{1+0.0016E_1}\text{(for } I=3\text{)} \quad \text{and} \quad \frac{E_2}{E_1}=\frac{1}{1+0.012E_1} \text{ (for } I=48 \text{ and } I=100\text{)}.$$

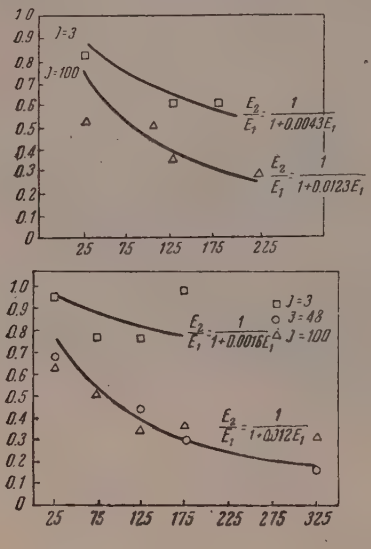

Fig. 1.

II. With a constant indirect stimulus and a constant photometric sensitivity of the eye E_1, one obtains:

$$\frac{E_2}{E_1}=\frac{1}{1+\beta\psi(x)}=\frac{1}{1+\beta\psi[x(I)]}. \qquad (7)$$

Formula (7) confirms the curves of Figure 1 from which it will be

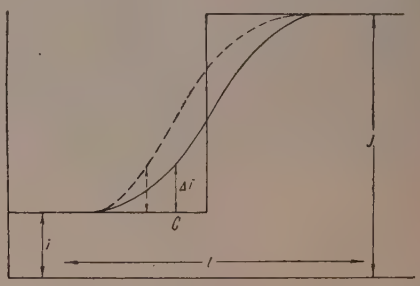

Fig. 2.

seen that with a E_1 constant for the given indirect stimulus the change in the photometric sensitivity of the eye depends upon the intensity of the direct stimulus and that this change tends towards a certain limit: while for the curves $I=3$ and $I=48$ a great difference is obtained, the curves for $I=48$ and $I=100$ practically coincide, whence it may be seen that the curve of relative change of the photometric sensitivity of the eye as dependent on the change in the intensity of the direct stimulus for I exceeding 48 units is approximately parallel to the abscissa.

III. With a constant photometric sensitivity of the eye and a constant intensity of direct stimulus we obtain

$$\frac{E_2}{E_1}=\frac{1}{1+\gamma\Delta_1(x)}. \qquad (8)$$

With increase in intensity of the indirect stimulus $\Delta_1 x$ is increased, and hence E_2 should decrease, which has actually been observed by

N. N. Dsidsischvili (Kravkov's laboratory, unpublished). According to these data, the photometric sensitivity of the eye is decreased with increase in the intensity of the indirect stimulus, at first rapidly and then gradually slowing down.

IV. With a constant sensitivity of the brain centres and a constant indirect stimulus, the ratio $\frac{E_2}{E_1}$ equals $\frac{1}{1+\alpha}$, where the constant α, independent of the intensity of the direct stimulus, is a magnitude equalling $\frac{\Delta_1 x}{\Delta x}$. This is supported by Kravkov's data, reported in his article «Effect of indirect light stimulation as a function of intensity of the direct stimulus», in which the values of this ratio have been determined for various intensities of the direct stimulus. For $I = 3.7$, 20 and 147 lux/white this ratio was found to equal 0.78, 0.73 and 0.75 respectively, i. e. it proved practically constant within the accuracy of readings.

Rather similar reasoning may be applied to explain the irradiation phenomena. If we have 2 adjacent objects, viz., a white one, reflecting $I^0/_0$, and a grey one, reflecting $i^0/_0$, the boundary between them will as a rule be seen not at the actual place of its existence, but shifted to a position C (Fig. 2). Assuming that this boundary is seen where $\frac{\Delta i}{i} = \Delta x \psi(x, l)$ and taking into account that upon exposure to the indirect stimulus we shall have $\frac{\Delta_1 i}{i} = \Delta x (1+\alpha) \psi(x, l)$, it can be shown that in the first case the change Δi per unit length

$$\frac{\Delta(\Delta i)}{\Delta l} = i \psi'(x, l) \Delta x,$$

while in the second

$$\frac{\Delta(\Delta_1 i)}{\Delta l} = i \psi'(x, l)(1+\alpha) \Delta x.$$

Hence

$$\frac{\Delta(\Delta_1 i)}{\Delta l} = \frac{\Delta(\Delta i)}{\Delta l}(1+\alpha), \qquad (9)$$

i. e. upon application of the indirect stimuli Δi seems to increase more rapidly, thus resulting in an apparent widening of the white band.

If the inversion phenomenon established for the absolute sensitivity of the eye is also accompanying the discrimination sensitivity, then it should be expected that in some cases the irradiation will not be increased, but on the contrary decreased, and that there exists a certain inversion point where the influence of the indirect stimulus will be zero. This conclusion is corroborated by the data by Bilezky and Kravkov on the effect of sound of various intensity on the irradiation effect in the eye.

It may thus be seen that all the changes in the discrimination sensitivity of the eye experimentally induced by application of the indirect stimuli can be reasonably explained by the assumption that the change in the threshold caused by the indirect stimulus does not depend upon the intensity of the direct stimulus and is determined only by the intensity of this indirect stimulus and the particular sensitivity of the brain, increasing with the enhancement of both these factors $\left(\alpha = \frac{\Delta_1 x}{\Delta x}\right)$. According to Kravkov[10], «the intensity of excitation induced by the indirect stimulus is a function of intensity of the direct stimulus». This thesis and all the inferences made therefrom result

firstly from the application of Weber-Fechner's law: $\frac{\Delta I}{I+\beta} = \Delta E = \text{const}$, which, in this form, by linking directly the sensation with the stimulus, is one of the most persisting scientific fallacies and, secondly, from an inadequate discrimination between the stimulus and the effect produced by the latter. This is why β, which has the dimension of the stimulus and can therefore be expressed in ergs, joules, calories, etc., is called by Kravkov the «additional excitation arising in the visual centres».

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BIOPHYSICS

ON THE NUMBER OF LIGHT SENSITIVE SUBSTANCES IN THE RETINA

By N. T. FEDOROV and V. I. FEDOROVA

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Colour vision is one of the fundamental problems of physiological optics. In spite of the great number of experimental studies and of theories of colour vision based upon the latter, the question is still open as to the number of light sensitive substances, the photochemical process in which gives rise to the impulses in the optical nerve, being thus the first stage of the perception of a definite colour.

While the founder of the thrichromatic theory of vision Thomas Young (1) postulates only the existence of «three kinds of nerve fibers», Helmholtz (2) (in his second post mortem edition of the «Handbuch der physiologischen Optik»)advances the hypothesis of three light sensitive substances of the eye. Helmholtz was followed by Lasarev (3), S. Hecht (4), E. Haschek (5) and others who have also admitted the existence in the eye of three different light sensitive substances with different absorption spectra. However, no conclusive experimental evidence has been brought forward by any of these authors which would have necessitated such an assumption. Moreover, Hecht's curves in particular (according to his own avowal) fail to explain the distribution of brightness actually observed in the spectrum of dichromates. As shown by Judd(6), these curves are incompatible with trichromatic mixture curves for spectral stimuli (the same holds true with regard to the curves once suggested by Lasarev).

As to the change in the hue in the course of chromatic adaptation of the eye, which led Haschek to the conclusion of the retina containing three light sensitive substances with different photochemical constants, the phenomenon in question can also be quantitatively accounted for by assuming the existence of a single light sensitive substance. A similar explanation was proposed by the author for the change in the hue caused by altering light intensity or by adding white light (7). An analysis of the dynamics of chromatic adaptation made Kravkov (8) assume the existence of several light sensitive substances with different constants of the photochemical and the reverse dark reaction. This conclusion should, however, be put down to the difficulties in obtaining reliable results by means of the methods applied by this author, since variation in brightness might have been disguised by changes in saturation or hue [cf. also Schober and Trilltsch (9)]. On the other hand,

by means of a very fine and laborious biochemical and spectrophotometrical analysis, G. Wald ([10]) succeeded in demonstrating the presence in the chick retina of only two light sensitive materials, viz., of the «visual purple» associated with the so-called scotopic rod vision, and of another violet substance which he called «iodopsin»; its absorption spectrum corresponds approximately to the photopic luminosity curve.

In the present study we have chosen another way to approach this problem. The brightness of the monochromatic light was compared under the conditions of purely foveal vision at the first moment of its action upon the eye, with the stationary brightness which is established after a prolonged exposure of the eye to this light. The difficulty of the experiments consisted in that such a chromatic adaptation of the eye was associated with some changes not only in brightness, but frequently also in the hue and saturation, the latter being subject to decrease.

Our purpose was always to achieve perfect matching and thus to avoid the changes in brightness to be disguised by those of saturation. We had therefore recourse to the following method. In the course of prolonged preliminary experiments the wave length, the hue of which was to be compared with that of the stationary field, and the amount of white light added to it have been selected in such a way that both fields appeared at the first instant identical. A quantitative analysis of this phenomenon of chromatic adaptation enabled us to gain a conclusive answer as to the number of light sensitive substances in the retina.

It follows from very accurate and reliable data of Wald ([11]) that the scotopic luminosity curve fully coincides with that for the number of quanta absorbed by the visual purple in various parts of the spectrum, that is to say:

$$B_\lambda = \alpha n = \alpha \frac{\varepsilon I_\lambda}{h\nu} X, \tag{1}$$

where α is a constant, n — number of quanta absorbed, ε — coefficient of light absorption, X — concentration of the light absorbing substance.

Having naturally assumed that the same correlation holds also for the photopic vision, it may be taken that, in order to equalize the brightness of both halves of the field of vision, an equal number of quanta should be absorbed at the given moment. Hence, in the simplest case when, in result of adaptation, neither the hue, nor the saturation do change, we write:

$$B_m = B_s, \tag{2}$$

where B_s — brightness of the field to which the eye was exposed for a long period of time, and B_m — brightness at the first moment of exposure of the eye to the light. Whence follows:

$$\alpha \frac{\varepsilon I_m}{h\nu} X = \alpha \frac{\varepsilon I_0}{h\nu} (X - x), \tag{3}$$

where X is the amount of the light sensitive substance decomposed by the light of I_0 intensity. In order to obtain x, we shall write the differential equation of the reversible photochemical reaction

$$\frac{dx}{dt} = k_1 \frac{\varepsilon I_0}{h\nu} (X - x) - k_2 x^n \tag{4}$$

and integrate it for the cases when $n = 1$ and $n = 2$. At $n = 1$

$$x = X \frac{k_1 \varepsilon I_0}{h\nu \left(\frac{k_1 \varepsilon I_0}{h\nu} + k_2 \right)} \left(1 - e^{-\left(\frac{k_1 \varepsilon I_0}{h\nu} + k_2 \right) t} \right), \tag{5}$$

whence we find for the stationary condition

$$x = X \frac{k_1 \varepsilon I_0}{h\nu \left(\frac{k_1 \varepsilon I_0}{h\nu} + k_2 \right)}.$$

By substituting this expression into the formula (3) we obtain:

$$\alpha \frac{\varepsilon I_m}{h\nu} X = \alpha k_2 \frac{\varepsilon I_0 X}{h\nu \left(\frac{k_1 \varepsilon I_0}{h\nu} + k_2 \right)}$$

or

$$B_m = B_s = k_2 \frac{B_0}{(k_1' B_0 + k_2')} = \frac{B_0}{aB_0 + b}, \qquad (6)$$

where

$$a = \frac{1}{\alpha k_2 X} \qquad \text{and} \qquad b = \frac{1}{\alpha}.$$

In those cases when in the course of adaptation a change occurs in the hue and saturation, we have to alter also the wave length in the field to be compared and to add to it white light. In this case we may assume that the number of absorbed quanta of the complex radiation will be

$$\frac{\varepsilon_1 I_m X}{h\nu_1} + X \int_{400}^{700} \frac{\varepsilon I_m}{h\nu} d\lambda = \frac{\varepsilon_1 I_m X}{h\nu_1} \left(1 + \frac{\int_{400}^{700} \frac{\varepsilon I_m}{h\nu} d\lambda}{\frac{\varepsilon_1 I_m}{h\nu_1}} \right) = \frac{\varepsilon_1 I_m X}{h\nu_1} (1 + \delta) = \gamma \frac{\varepsilon_1 I_m X}{h\nu_1} = n\gamma, \quad (7)$$

i. e. B_m will equal $\alpha n\gamma = \alpha' n$, whereas in the equation (6) only the coefficients a and b will be changed.

For $n = 2$ the integral of the equation (4) is as follows:

$$x = \frac{2X}{1 + \frac{1 + e^{-\frac{k_1}{\alpha} Bt\beta}}{1 - e^{-\frac{k_1}{\alpha} Bt\beta}} \cdot \beta} \qquad (8)$$

where

$$\beta = \sqrt{1 + \frac{4b}{B}}, \quad b = \frac{k_2 X^2 \alpha}{k_1} \text{ and } B = \alpha \frac{\varepsilon I}{h\nu} X.$$

For the stationary condition the following ratio will be derived:

$$x = \frac{2X}{1 + \beta} \qquad (9)$$

and

$$X - x = X \frac{\beta - 1}{\beta + 1}, \text{ while } \beta = \sqrt{1 + \frac{4b}{B_0}}.$$

[It can be readily shown that x determined after formula (9) is identical to x computed from the equation for the stationary state $\left(\frac{dx}{dt} = 0\right)$:

$$x = \frac{X}{2} [(b_1^2 B^2 + 4 b_1 B)^{\frac{1}{2}} - b_1 B], \text{ where } b_1 = \frac{k_1}{k_2 X \alpha} = \frac{X}{b},$$

which is used by Hecht himself].

By means of a reasoning which is quite similar to that applied to the foregoing case, we obtain ultimately from formulae (2), (3) and (7):

$$B_m = B_s = aB_0 \frac{\sqrt{1+\frac{4b}{B_0}}-1}{\sqrt{1+\frac{4b}{B_0}}+1}. \tag{10}$$

Formulae (6) and (10) concern only the case when in the eye there is present a single light sensitive substance. If however we postulate, following Helmholtz, the existence of three different chemical substances, we should write, instead of formulae (6) and (10):

$$B_m = \sum_{i=1}^{i=3} \frac{B_{0i}}{a_i' B_{0i} + b_i'}, \tag{11}$$

$$B_m = \sum_{i=1}^{i=3} a_i B_{0i} \frac{\sqrt{1+\frac{4b_i'}{B_{0i}}}-1}{\sqrt{1+\frac{4b_i'}{B_{0i}}}+1}. \tag{12}$$

It follows, besides, from the law of brightness additivity that

$$B_0 = \sum_{i=1}^{i=3} B_{0i}. \tag{13}$$

From formulae (6) and (10) it may be seen that whatever be the wave lengths chosen on the assumption of a single light sensitive substance characterized by the constants k_1 and k_2, the fields will remain equally bright after the stationary sensitivity level will have been attained as a result of adaptation of the eye to the given stimulus (provided they were so prior to the beginning of adaptation).

If, however, in the cones there are contained three different substances, then the constants k_1 and k_2 will be different too, and, upon adaptation to radiations of various wave lengths λ', λ'' and λ''', though the original brightnesses of these radiations are identical, i. e.

$$B_{01}' + B_{02}' + B_{03}' = B_{01}'' + B_{02}'' + B_{03}'' = B_{01}''' + B_{02}''' + B_{03}''',$$

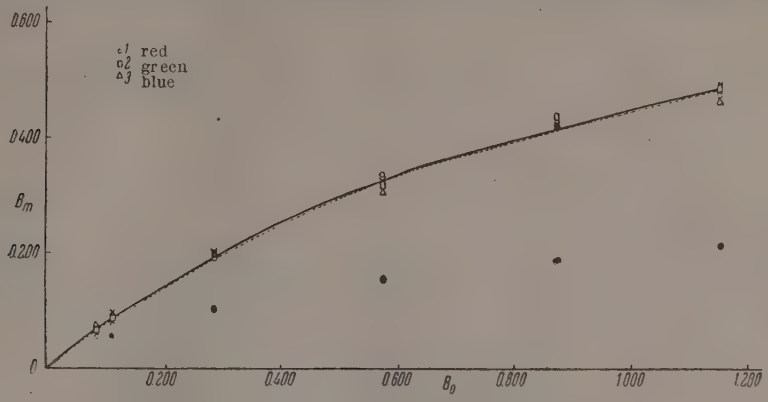
owing to the differences in the coefficients k_1 and k_2, their stationary brightnesses B', B'' and B''', as determined after formulae (11) and (12), will necessarily be unequal.

Experimental Results

Direct Method. Having closed the one half of the field (field's size 1.5°) of the Helmholtz' colour mixture apparatus, the experimenter (V. I. Fedorova), after preliminary dark adaptation, was adapting herself during 90 sec.* to the light of the other half with a wave length of, say, 660 mμ. Thereupon the first half was opened for a very short time ($\sim$0.5 sec.), and the experimenter was asked whether or not it was equal in colour

* In full agreement with the data by Kravkov, Schober and Triltsch, as well as those by Allmack, our results showed that the stationary level is attained in 90 sec.

and brightness to the second half. By means of numerous preliminary tests, such parameters of the first half were found which made it identical to the second one. The experiments were then repeated with



——— calcul. after f. (10); — — — — calcul. after (6); x x x — calcul. after (14) for $\lambda = 660$ mμ; ○ ○ ○ calcul. after (14) for $\lambda = 460$ mμ.

another and a third wave length ($\lambda_2 = 510$ mμ, $\lambda_3 = 460$mμ). Equal initial brightnesses B_0 were used according to the spectral luminosity curve, which had been preliminarily determined for V. F. by the step-by-step method. The whole series was then repeated on another level of initial brightness. The results of a number of such measurements for six brightnesses $B_0 = 1.16$; 0.88; 0.58; 0.29; 0.116 and 0.088 obtained after prolonged training are given in the table and the figure where B_0 is plotted along the abscissa and the corresponding B_m is plotted along the ordinate. The solid line is computed after the formula:

$$B_m = 1.09\, B_0 \frac{\sqrt{1 + \frac{4.37}{B_0}} - 1}{\sqrt{1 + \frac{4.37}{B_0}} + 1}$$

and the dotted one after the formula:

$$B_m = \frac{B_0}{B_0 + 1.23}.$$

It may thus be seen that within the accuracy of the method all the points for all the three colours are fitting within one and the same curve. It may also be seen that this phenomenon of chromatic adaptation permits of no determination of the order of the reverse reaction. It follows from the curves of fundamental responses for the eye of Fedorova, which enabled us to explain a number of facts of physiological optics, that the radiations 660 mμ and 510 mμ are composed only of two fundamental responses R and G, that for 660 being $B_r : B_g = 4.91$, and for 510 mμ $B_r : B_g = 0.168$. Hence, if we assume the existence in the eye of several light sensitive substances, we should obtain for these radiations B_m equal to

$$B_m = \frac{B_r}{a_1 B_r + b_1} + \frac{B_g}{a_2 B_g + b_2} \qquad (14)$$

and to the expression derived from formula (12) for the case of a binocular dark reaction. An analysis of the experimental data shows that,

if $a_1 = 1.26$, $b_1 = 1.03$, $a_2 = 5.7$ and $b_2 = 2.3$, the value of B_m computed from formula (14) proves in good agreement with the actually observed ones (see table and figure).

B_0	B_m (460 mμ)	B_m (510 mμ)	B_m (660 mμ)	B_m (form. 10)	B_m (form. 6)	B_m (form. 14) for $\lambda = 660$ mμ	B_m (form. 14) for $\lambda = 510$ mμ
1.16	0.463	0.480	0.472	0.482	0.479	0.486	0.209
0.88	0.427	0.434	0.417	0.413	0.410	0.422	0.187
0.58	0.309	0.322	0.335	0.316	0.316	0.328	0.152
0.29	0.200	0.195	0.193	0.193	0.188	0.200	0.100
0.116	0.092	0.084	0.077	0.092	0.084	0.093	0.050
0.088	0.0785	0.070	0.064	0.072	0.063	0.074	0.038

As to the B_m values computed for $\lambda = 510$ mμ with the same constants a_1, b_1, a_2 and b_2, they are represented in the figure by crosses and tabulated in the last column of the table. It will be seen that the second alternative (existence in the eye of several light sensitive substances) is to be discarded. Similar conclusions are arrived at by means of indirect evidence.

Indirect Methods. It follows from our experiments that chromatic adaptation does not change the heterochromous brightness matching of red, green and blue fields with the achromatic one. It was found further on (experimenter V. I. Fedorova), that if one becomes adapted to a field, one half of which is chromatic (λ_1), and the other (of equal brightness B_0) achromatic, and if as soon as the stationary level is attained, the achromatic field is substituted by the chromatic one (of the same λ_1) of brightness B_0, identity will be obtained merely by changing λ_1 for $\lambda_1 + \Delta\lambda_1$ or by adding white light, both without changing brightness B_0. This could not take place, if we had to deal with several light sensitive substances.

Thus, the results of both direct and indirect experiments lead us to the conclusion that the cones of the eye contain but one light sensitive substance. The mechanism underlying chromatic vision should thus be sought for in the existence of nerve fibers which connect the retina with three different centres of the brain.

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CHEMISTRY

A NEW METHOD OF PRODUCTION OF POTASSIUM SALPETER BY EXCHANGE DECOMPOSITION OF POTASSIUM CHLORIDE WITH NITRIC ACID AT LOW HNO_3 CONCENTRATIONS AND TEMPERATURES

By V. I. NIKOLAEV and A. G. KOGAN

(Communicated by N. S. Kurnakov, Member of the Academy, 29. XI. 1938)

Without touching here upon the theoretical principles of the method suggested, which was done elsewhere(1), the authors are giving in the present paper mainly the results of additional investigations concerning the «tails» of the procedure of potassium salpeter production, and suggest a new variant of utilizing these «tails».

The course of the procedures for the recovery of KNO_3 is as follows.

1. In a 37.7% nitric acid potassium chloride is introduced in such a quantity that the weight ratio $\frac{HNO_3(37.7\%)}{KCl}$ be equal to 6.0 units.

The solution obtained at +44° is cooled to +°1, +2°. 29.48% KNO_3 of the theoretical yield are isolated.

2. After the separation of KNO_3 the mother liquor is resaturated with potassium chloride at +44°. The solution thus formed being cooled to +1°, +2°, 24.82% more of the theoretical quantity of KNO_3 are isolated.

3. To the second mother liquor 36.8% of water are added, and the solution is saturated with potassium chloride at +44°. Being cooled again, this time only to +8°, 22.4% more of pure salpeter are isolated.

Thus, in the course of three subsequent dissolutions of KCl and coolings of the solutions to +1°, +2°, 76.7% of pure salpeter out of the theoretically possible quantity of KNO_3 are isolated.

4. From a separate study of the vacuum-effect on the distribution of hydrochloric acid and nitric acid in individual fractions of the distillate (figure) one might expect an almost complete separation of the hydrochloric acid from the nitric one in the first fractions with a vacuum of 100 mm mercury.

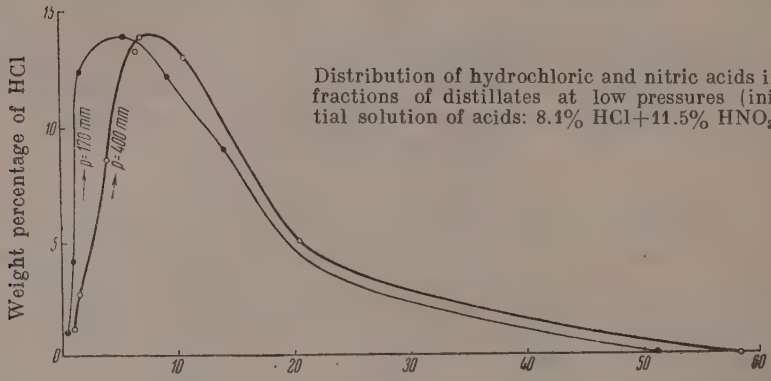
It proved, therefore, possible to propose the following methods for utilizing the mother liquor «tails» left behind after the above described procedures to obtain 76% of pure salpeter.

The last mother liquor is saturated with an equivalent of potassium chloride calculated so as to bind all of the free nitric acid (the composition of the mother liquor may be represented as follows: HNO_3=2.07%, HCl= =16.36% and KNO_3=9.25%).

At a 90—100 mm vacuum a quarter (by weight) of the acid mixture is distilled from the mother liquor, HNO_3 constituting only 0.75% of the

mixture. The distillation temperature is being maintained at about 60°. The residue is cooled down to +1°, +2°. A mixture $KCl+KNO_3$ is formed. Similar operations are repeated four times more with each fresh mother liquor, i. e. after saturation with a quantity of potassium chloride equivalent to the nitric acid in the mother liquor (with the above-mentioned recalculation of the mother liquor composition), and after the distillation of the acids in a quantity making up one quarter of the mother liquor by weight, the residue of the distillate is cooled down to +1°, +2°, and the $KCl+KNO_3$ mixtures are isolated.

5. All the mixtures obtained are used for a new loading, with nitric acid instead of potassium chloride alone. In the new cycle the admixture



Distribution of hydrochloric and nitric acids in fractions of distillates at low pressures (initial solution of acids: 8.1% HCl+11.5% HNO_3)

Weight percentage of HNO_3 in fractions of distillates at 400 and 170 mm vacuum.

of KNO_3 to potassium chloride causes: a) a higher yield than 76% of pure salpeter; b) a smaller quantity of nitric acid in a repeated cycle: nitric acid (37.7%) should be introduced in a quantity not exceeding that which secures the ratio $\frac{HNO_3(37.7\%)}{KCl} = 6.0$.

Thus, the second cycle is more perfect than the first one. In the first cycle, however, the total loss of nitric acid amounts to about 3.4%, since the salpeter formed in the $KCl+KNO_3$ mixtures is regenerated in the second and the subsequent cycles, increasing the total yield of pure salpeter.

N o t e. The graph (figure) shows that if we use a higher vacuum (say, 50 mm mercury) when distilling mixtures of $HCl+HNO_3$ acids, the separation of the hydrochloric acid from the nitric one will be still more perfect, and the total loss in nitric acid will be still reduced.

6. The 13—17% hydrochloric acid, contained in separate distillates, proves to be quite free from nitrogen oxides and of a concentration from 18 to 20% and above, after an addition of about 40% of $MgCl_2.6H_2O$, and a distillation and blowing through of a slight air stream.

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PHYSICAL CHEMISTRY

MECHANICAL PROPERTIES OF «TWO-DIMENSIONAL» CAOUTCHOUC. THE MICELLAR STRUCTURE OF CAOUTCHOUC

By A. A. TRAPEZNIKOV

(Communicated by A. N. Frumkin, Member of the Academy, 28; XI; 1938)

It has been suggested by a number of authors ([1],[2]) that the enhancement in the mechanical properties of caoutchouc during the filling of it with active fillers is connected with the transition of the caoutchouc into a special state on the surface of the filler particles. Rehbinder and Margaretov ([3]) studying the mechanism of activation due to the fillers, consider this change in the mechanical properties as a transition of the caoutchouc into a two-dimensional film-state of solvate membranes which, similar to thin liquid layers ([4]), possess anomalous mechanical properties (elasticity in shear), as well as a peculiar colloidal structure.

In the present investigation the mechanical properties of thin films of caoutchouc were studied with the aim of discovering their anomalies, the appearance of which may be expected for sufficiently thin and, most probably, oriented films.

The mechanical properties, characterized by deformation curves during stretching, were studied with an apparatus consisting of a paraffined trough containing water, on the surface of which was placed a film of caoutchouc, spring balances (glass needle) to measure the tension, and two barriers to stretch the film. One barrier in the form of a paraffined strip of paper, floating freely, was joined by steel hooks to the lower ends of the balance; the other, a glass one, moved along the ends of the trough.

The films were formed by the evaporation of a drop of a toluene sol. However, preliminarily, a layer of palmitic acid* was deposited onto the water surface which prevented: 1) the unbounded spreading of the drop of the sol over the whole water surface, and 2) the adhesion of the film to the edges of the vessel (when, in order to obtain very thick films, the vessel employed was of small dimensions). The areas of spreading, determining the thickness of the film (from the weight of caoutchouc contained in one drop), may be controlled by changing the surface concentration of the layer of palmitic acid, which becomes more dense during the spreading of a sol drop up to about a condensed state.

The tension was calculated with reference to 1 cm^2 of the initial cross-section. The rubber was purified by precipitation with alcohol from a toluene solution, preliminarily filtered from pectinic substances.

* Palmitic acid does not affect the mechanical properties of the caoutchouc films (in these experiments); the form of a sol drop, which scarcely changes over a long period of time, shows that the palmitic acid does not penetrate into it.

In such a manner there were obtained films ranging from 25 to 4000 Å. For thicknesses greater than 500—800 Å. the films are coloured due to interference, while for smaller thicknesses, they are colourless and visible only in reflected light against a dark background. These thin films are porous (veined), which accounts for the comparatively low rupture tensions.

In Fig. 1 are shown deformation curves for the stretching of caoutchouc films (light crepe) on distilled water for thicknesses ranging from 25 to 3000 Å. It may be seen that the films are divided into two groups: «thick» films, from 300 Å and higher, and «thin» films, up to 200 Å. The «thick» films give deformation curves similar to those obtained in the case of stretching of three-dimensional volume caoutchouc; they are S-shaped and are characterized by a section

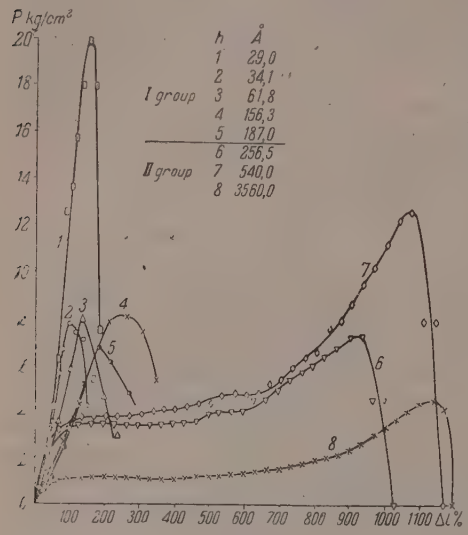

Fig. 1.

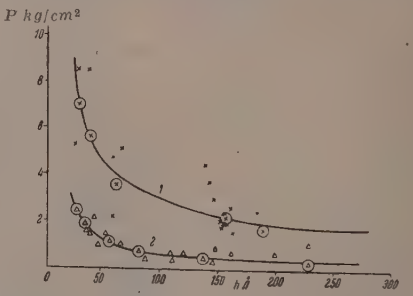

Fig. 2. *1*—light crepe on distilled water, *2*—smoked sheet on tap water (Δl=66%).

of plastic elongation about 500—600% (the total rupture elongation is 1000—1200%). The «thin» films, on the contrary, lack plasticity, their deformation curves being almost linear, i. e., the connexion between deformation and elongation for this group of films is in close conformity with Hooke's law. In compliance with this the curves characterizing a drop in the load during contraction of the stretched «thin» films give almost no residual deformation or hysteresis loop, while the hysteresis loop formed by the curves of the contraction of «thick» films is very large and is determined mainly by the amount of plastic elongation. As the films become thinner, the modulus of elasticity increases as is shown in Fig. 2 and complete elongation at the rupture decreases from 1000 to 200—300%.

These data corroborate the above assumption concerning the appearance of anomalously high mechanical properties for very thin caoutchouc films.

An especially sharp qualitative distinction in the mechanical properties is observed in films beginning with a thickness of 200 Å. Upon further thinning of the film, the character of the curve does not change except its becoming more steep, i. e. the modulus increases. The appearance of visible porosity for films of these thicknesses (starting with 200 Å) points to the impossibility of a further homogeneous thinning of the films and to the fact that at these thicknesses a minimum thickness is attained which seems to correspond to the size of a structural unity, for example, to a caoutchouc micella.

It follows hence that the difference in the mechanical properties of the two groups of films may be explained in the following manner. In the thick films, the caoutchouc micellae after evaporation of the sol-

vent remain in a state of chaotic interweavement and the plastic region of expansion corresponds to the slip and gradual orientation of micellae. However, in thin films, owing to the drop being spread over a large area, the micellae arrange themselves in one layer with a definite orientation, which excludes their plastic slip past one another. As the thickness of the film descreases, the orientation conditions become better, which leads to an increase in the modulus of elasticity. Two-dimensional caoutchouc is equivalent to three-dimensional «crystallized» caoutchouc.

Owing to the porosity of the thin films, their actual thickness is somewhat greater than the average one and it may be determined by condensing the film (by eliminating interspaces), for example by means of slight compression

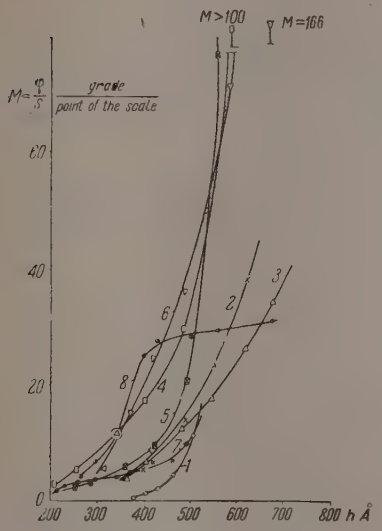

Fig. 3.

Light crepe

Number of curve	Sol concentration	Number of drops induced on the surface	Lining
1	0.078	42	Water
2	0.078	60	0.001 *n* HCl
3	0.078	60	0.001 *n* HCl
4	0.0195	240	0.001 *n* HCl
5	0.078+5% of palmitic acid	55	0.001 *n* HCl
6	0.156+2.5% of palmitic acid	30	0.001 *n* HCl
7	0.078+5.2% of cetylic alcohol	55	Water
	synthetic caoutchouc		
8	0.127	40	0.001 *n* HCl

by blowing. However, from solutions of very small concentrations of caoutchouc, complete films are not obtained, but after evaporation separate unconnected islets remain. By bringing together and condensing such islets, a value of the limiting thickness also may be obtained. The value $h=360$ Å was obtained by this method of blowing, for sol concentrations from 1.42 to 0.07%.

For high concentrations (0.5—1.42%) the values of h may lack accuracy because of incompletely uniform spreading of concentrated solutions; but the fact that for dilute solutions (even to C=0.02%) homogeneous disintegration (division into small parts) of micellae does not take place, is corroborated by the data of the measurement of the modulus of elasticity for shear by the method of twisting a disc in the film which is suspended on an elastic thread ([5]). In Fig. 3 are given curves of the dependence of the modulus (in degrees of revolution of the upper end of the thread per one division of the scale during the shift of the light beam) upon the thickness of the film during its compression (condensing of the islets) by the barrier on the surface of the water in the trough. All the solutions, from 0.15 to 0.02%, in spite of the incomplete homogeneity of the films, give a sharp increase in the modulus at a thickness of about 400—500 Å (Curves *1*, *2*, *3* and *4*). The introduction of surface-active substances, for example palmitic acid

(measured on 0.001 n HCl), into the caoutchouc sol, which accelerates the spreading of a drop and aids the disintegration of the film into separate micellae, does not change the limiting values of the film thicknesses (Curves *5*, *6* and *7*) lying from 350—500 Å. This undoubtedly points to the fact that this value of the thickness is not accidental. Palmitic acid (2.5 to 5.0%) which aids in making the films homogeneous, enhances its mechanical properties. Synthetic caoutchouc gives films having a thickness of the order of 300—350 Å.

The values of the thicknesses, h=400—500 Å, may apparently be attributed to the length of the chains in a micella. According to Hengstenberg's[6] exograms, crystallites in stretched caoutchouc possess a length, width and thickness of the order of 600, 500 and 150 Å respectively.

Preliminary data obtained with fractions of caoutchouc prepared by a method of fractional precipitation with alcohol (in prof. Lipatov's laboratory) showed that the film thickness of the first fraction is approximately 700 Å, and that of the second fraction approximately 400—500 Å. Thus, the indicated value is the mean value for a mixture of polymer homologues (the third fraction was not investigated).

These data may be considered as a result of the micellar structure of caoutchouc in solution. For isolated macromolecules the film thickness, in a compressed condition, should have amounted to either 4000—8000 Å for vertical orientation, or to 2.5 Å for plane location of the chain. The film could have been homogeneously thinned to thicknesses considerably less than 200—400 Å. However, the assumption of any arbitrary average distribution (interweaving) of chains, giving a thickness of 500 Å for solutions of various concentrations (in the region sol solution at C$<$0.1—0.24%) and spreading with a different speed, is but very slightly probable. However, thicknesses of the order of 2.5—3.0 Å may be obtained only under the influence of a considerable stretching force, for example by a high two-dimensional pressure arising on the surface of mercury. Films on mercury, after evaporation of the toluene, continuously spread to a thickness of 2.5 to 3.0 Å, below which the film loses coherence (qualitative experiments of the adhesion of a film to a rod). The loss of coherence at 2.5—3.0 Å evidently corresponds to a gaseous structure of the monolayer with the disintegration of micellae to separate, flatly lying not touching chains. This agrees with Keenan's results (7). The results of an electron-diffraction analysis of thin ($<10^{-5}$ cm) films of caoutchouc on water (8,9) show that they are oriented (crystalline).

The method of monolayers may be adapted to colloids in spite of the formation of islets, since at corresponding conditions the structure of the given islets is sufficiently definite; they are two-dimensional micellar aggregates, formed during the process of evaporation of the solvent.

This investigation, which was started in the Laboratories of the Liebknecht State Pedagogical Institute, is being continued in the Department of Physical Chemistry of Disperse Systems of the Colloidal Electrochemical Institute of the Academy of Sciences of the USSR.

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GEOCHEMISTRY

ISOMORPHIC SUBSTITUTIONS IN APATITE

By DUNCAN McCONNELL

(Communicated by A. E. Fersman, Member of the Academy, 10. XII. 1938)

Unfortunately some of the results published by me ([1],[2]), as well as a collaboration with Dr. Gruner([3]), have been misinterpreted recently in this journal([4]). Whatever the source of the misunderstanding it seems essential that I make my contentions quite clear, and point out certain weaknesses in the arguments used in the attempt to discredit portions of my earlier conclusions.

Borneman-Starinkevitch attacks the hypothesis which postulates carbon replacing calcium in the apatite structure, although this type of substitution has been demonstrated in terms of three published analyses: ellestadite([2]), francolite ([4]) and dahllite([5]). Another more recent analysis of francolite from Devon also demonstrates this type of substitution and these results will appear later. Obviously the accuracy of these analyses cannot be demonstrated in any material way, but it is well to remember that three different analysts are involved. Anybody familiar with the names of American chemists will recognize the experience and skill to be associated with these names:

for ellestadite—Dr. R. B. Ellestad, Laboratory for Rock Analysis, University of Minnesota;

for francolite—Dr. R. B. Ellestad;

for dahllite—Ledoux and Company, New York, N. Y.;

for francolite—Dr. E. B. Sandell, Department of Analytical Chemistry, University of Minnesota.

Furthermore, it was found in our work([3]) that the carbon atoms formed CO_3-groups which had dimensions almost identical with those found for these groups in the structures of calcite and aragonite.

Borneman-Starinkevitch has attacked these rather precise measurements by stating that carbon is not known as a cation. Now according to the accepted definition, a cation is an ion which migrates to the cathode during electrolysis. Quite obviously the term «cation» has no meaning when discussing the atoms (or ions) within a crystal lattice, and the fact that carbon is unknown as a cation has no bearing on our conclusions.

In order to account for the CO_2 which is in excess of the amount which can be assigned to phosphorus positions of the lattice, Borneman-Starinkevitch has concluded that an aqueous solution of carbonic acid is present in these minerals. However, no evidence to substantiate this contention has been furnished. It would seem most peculiar that this should be true in all four of the cases cited, particularly, in view of the fact that the para-

genesis of each of these minerals is quite distinct. Furthermore, the textures are different. Dahllite and francolite are fibrous but ellestadite is not fibrous.

In addition to the evidence obtained for minerals, the substitution of carbon for calcium has been demonstrated in another collaboration on tooth-substance ([6]). Four analyses are given in this work and these determinations involve a fourth analyst, namely, Dr. W. D. Armstrong, Department of Physiological Chemistry, University of Minnesota.

Borneman-Starinkevitch states: «... the existence of CO_4^{4-} ions is also unlikely» in discussing the results which Dr. Gruner and I obtained. I wish to be emphatic in denying that we ever suggested the existence of CO_4^{4-} ions. I have described a CO_4-group in several of my own works as well as in the collaboration with Dr. Gruner. There is a very clear-cut distinction between a CO_4^{4-} ion and a CO_4-group. The former could exist only in an electrolytic solution (were it to exist at all) while the latter was postulated for a crystal lattice. In distinguishing between a CO_4^{4-} ion and a CO_4-group it must be pointed out further that no charge of whatever magnitude can be assigned to our CO_4-group. One can only speak of coordination number in connection with a crystal lattice and, consequently, the assignment of a charge to our CO_4-group (as has been done by Borneman-Starinkevitch) completely changes the meaning of our conclusions.

There are hydroxyl groups and fluorine atoms available and they may well be associated with those tetrahedra which contain carbon at their centers, as suggested by Borneman-Starinkevitch, but at best this is a supposition because there are no sufficient data to justify the assignment of the hydroxyl groups and fluorine atoms to certain oxygen positions in preference to other oxygen positions within a structure of this complexity.

Borneman-Starinkevitch postulates the use of the hypothetical molecule $3CaCO_3 \cdot 2Ca(F,OH)_2 = Ca_5C_3O_9(F,OH)_1$ to account for francolite. Dr. Gruner and I have already shown that the tetrahedral configuration for carbon is not the most stable configuration and probably does not exist except to a limited extent in special types of compounds. In assuming the existence of this molecule it is necessary that one also assume that a compound exists in which all of the tetrahedra have carbon atoms at their centers. The second assumption is imperative if one is to obtain a compound which has the apatite structure, and the compound must have the apatite structure if it is to be isomorphic with other apatite molecules.

If our conclusions are not valid, they must be attacked through the presentation of new experimental data or through the reinterpretation of our measurements in accordance with fundamental precepts.

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* In the manuscript is given erroneously 1938. *Edit.*
** In the manuscript is given erroneously 1936. *Edit.*

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GEOCHEMISTRY

ON SOME ISOMORPHIC SUBSTITUTIONS IN THE APATITE GROUP

By N. V. BELOV

(Communicated by A. E. Fersman, Member of the Academy, 8. XII. 1938)

Far from seeing any particular difference between the results obtained by McConnel and Borneman-Starinkevitch, I am rather inclined to regard the tables of the latter with Si and S-apatites, and especially with Na and TR, as a valuable complement to the tables of the former, considering the rigorism of Borneman who is avoiding commas within the brackets, and prefers the classical conception of the end-components in isomorphic series. Essential, however, is the discord in the opinions as to the possibility of mutual substitution between carbon and calcium, on the one hand, and carbon and phosphorus, on the other.

I wish to be as emphatic as McConnel, yet in maintaining that the CO_3 and CO_4-groups (without predetermining the existence of the latter), in which the tetravalent cation C^{4+} is lying among three or four bivalent O^{2-} ions respectively, are always anions $(CO_3)^{2-}$ and $(CO_4)^{4-}$, no matter where the group has been found, in a solution or in a lattice. The question is of importance whether the formation of such a group is possible crystallochemically.

The experience acquired in teaching taxonomy of crystalline structures shows that the best method of representing these are Pauling's polyhedra. The latter are especially appropriate when essential structural features are to be grasped, i. e. in the case of first, yet still sufficient, approximation.

In Pauling's polyhedra the structure of apatite $Ca_5(PO_4)_3 \vdots F = (CaI)_2 \vdots (CaII)_3 P_3O_{12}F$ is given in Figs. 1, 2, 3, 4, with the addition of a scheme (Fig. 5) of the space group of apatite o C^2_{6h}. For two kinds of cations Ca (first approximation) the coordination polyhedra are the same: straight trihedral prisms. The prisms of the cations CaII in the form of triple stars are placed along the sixfold screw axis (6_3) in such a way that one edge of each prism (common for three prisms) coincide with this axis. Along the axis the three prisms rotate through 60° for each half-period with respect to height so that none of the prisms has a common face, nor a common base with another. The combination of the prisms placed along the sixfold axes may be regarded (Fig. 1) as an endless linear cation $(Ca_3F)_\infty$. The ions of the halogen are lying on the axis of cation. The prisms of the cation CaI are also drawn out in endless linear cations—columns, but along threefold axes of the C^2_{6h} group, being placed singly at each level of the column in such a way that the lower base of the upper prism is always the upper base of the underlying prism (Fig. 2). Two kinds of columns of the prisms CaI and CaII are inter-

connected by columns of discrete PO_4 tetrahedra. These columns are given in Fig. 3. Through the middle of such a column a twofold screw axis is passing (2_1). Quite similar columns of discrete SiO_4 tetrahedra are characteristic of the olivine Mg_2SiO_4, zircon $ZrSiO_4$, kyanite Al_2SiO_5, as well as $CaSO_4$, $BaSO_4$, etc. The distribution of the O ions not at two but at four levels in these connecting columns causes the prisms in the columns CaI and CaII to be placed at different levels: the CaI ions are 1/4 of the period higher (or lower) than the CaII ions.

The whole scheme of the apatite structure is shown in Fig. 4, where it may be easily seen to be a kind of a graphical representation of the signs of the scheme of the space group C^2_{6h} (Fig. 5).

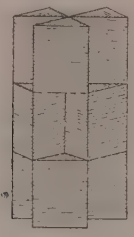

Fig. 1. Cations Ca II.

Fig. 2. Cations Ca I.

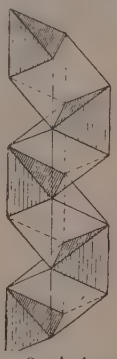

Fig. 3. Anions PO_4.

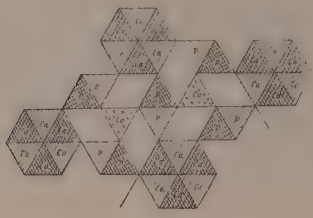

Fig. 4.

Fig. 5. Space group C^2_{6h}. Symmetry center at axes 6_3 and 2_1 in O and $^1/_2$C. Horizontal planes of symmetry at the heights $^1/_4$ and $^3/_4$ C.

McConnell is quite right in maintaining that it is impossible (simply from steric considerations) to introduce the CO_3-group instead of the halogen on the sixfold axis and, on the other hand, it seems desirable to introduce the cations C^{4+} into the already formed triads of O ions along the threefold axes, where these triads are the bases of prisms, enclosing cations CaI. However, with such a distribution of the CO_3-groups (and McConnell fails to take this into account) two essentially different possibilities should be considered. If all the CaI be substituted for C, i. e. all the prisms remain empty and all the bases be filled with carbon, which corresponds (the previous lattice symmetry being preserved) to the substitution $4Ca^{2+} \rightarrow 4C^{4+}$, then everything is all right from the crystallochemical viewpoint, provided due compensation for the increase of the total charges by a corresponding reduction of the + charges in other parts of the lattice take place.

But McConnell himself says that a substitution of carbon for calcium is possible only on a limited scale, following statistical probability, or at random. It may be easily seen that in this case only such substitutions are

possible, as when one cation C^{4+} is substituted at once for two cations Ca^{2+}, i. e. 1 $Ca^{2+} \rightarrow \frac{1}{2} C^{4+}$. As a matter of fact, Pauling's principles with their simple electrostatic logic do not allow the cation C^{4+} to be placed at the prism face, occupied by another cation. With an arrangement of the prisms characteristic of CaI, when the upper base of one prism is the lower base of another, the entrance of the cation C in any base causes both prisms to become empty, i. e. with a statistical substitution of carbon for calcium, one cation C^{4+} is necessarily substituted for two cations Ca^{2+}. (Such is, in particular, the structure of calcite, wherein cations are everywhere arranged within the common face of two empty octahedra.)

This circumstance is upsetting the arrangements of cations in 42 positions of the apatite unit cell so accurately balanced by McConnell. Apart from this it has been found that the introduction of 1 $Ca^{2+} \rightarrow \frac{1}{2} C^{4+}$ enables the cations in both cases treated in detail by McConnell—those of francolite and dahllite—to be balanced with equal precision. Namely, if we take 1.004 instead of McConnell's normalizing factor 0.9978, we obtain for francolite:

$$10\ Ca = 9.750\ Ca + 0.076\ Mg + 2 \times 0.087\ C$$
$$6\ P = 5.289\ P + 0.026\ V + 0.684\ C.$$

For dahllite we substitute 1.0304 for McConnell's factor 1.0245 and we obtain:

$$10\ Ca = 9.20\ Ca + 0.23\ Na + 0.06\ K + 2 \times 0.255\ C$$
$$6\ P = 5.27\ P + 0.01\ S + 0.73\ C\ (= 6.01\ P).$$

In the latter case $Al_2O_3 + SiO_2$ become unnecessary, the author himself regarding these as being determined by argillaceous admixtures.

Thus, the above statement as to the method of substitution of C^{4+} for Ca^{2+} results in a highly satisfactory balance of cations.

At what cost is, however, such a balance attained, has not been shown by McConnell. After all the figures of the analysis except oxygen (which in all cases must supplement the decreased total number of the ions to 42) are divided by the above-mentioned factors, there takes place a disturbance of the balance of + and — charges, which is a fundamental balance of any analysis. In the analysis of francolite McConnell shows : 49.574+ and 49.732—.But in dahllite, where the normalizing factor is high, the divergence is already 51.343+ and 49.817—, and McConnell has passed over these figures, while the inconsistency of such a divergence is obvious. Since no better agreement is observed in the supposed modified balance, we naturally reach the conclusion that the statement made by McConnell as to the possibility of a substitution of carbon for calcium in the isomorphous apatite series remains as yet unproved.

From the above it is clear that the doubts which have arisen refer not to the figures of the analyses (the accuracy of which is guaranteed by the authority of the American analysts), but rather to their arbitrary modification by normalizing factors, which makes the modified figures contradictory.

Such is not the case with the possibility of substitution of carbon for phosphorus in the PO_4-group. It should at once be noted that such a substitution does not necessarily mean that the fourfold coordination must be assigned to the ion C^{4+}. If corresponding distances in apatite are taken, it is found that the P^{5+} ion in the PO_4-group more closely approaches the three O ions (1.49; 1.60 and 1.60 Å) than the fourth (1.82 Å). The P^{5+} ion is, as it were buried (on account of its smallness) in one of the bases of the tetrahedron. In introducing into the tetrahedron instead of C^{4+} a still smaller one, we are evidently burying it still deeper in the base of the tetrahedron; according to

Coulomb's law the fourth distance is respectively increased and we are entitled to speak of a group or an anion $(CO_3)^{2-}$. Supposing the fourth apex to be occupied by F^{1-} or $(OH)^{1-}$ instead of O^{2-}, and compensating in this way the substitution of C^{4+} for P^{5+}, we obtain the formula $Ca_5C_3O_9$ (OH, F)$_4$ postulated by Borneman for one of the terminal components of the isomorphous apatite series. Such a degradation of the tetrahedra—a decomposition of these to a triangle + an apex not combined with the latter—weakens the significance (see the scheme) of the column of tetrahedra as a connecting link between the cation columns, and indeed we find McConnell to point out the fibrous nature of the CO_2-apatites—francolite and dahllite—in contradistinction to the other members of the isomorphous series.

The CO_3-triangle plane proves thereby to be not perpendicular but inclined to the threefold axis which moreover does not pass through the triangle. A perfectly similar arrangement of the CO_3-group we encounter in the structures of other also trigonal fluocarbonates—bastnesite and parisite.

The considerations referred to above are corroborated by the structure of analogous BO_3-groups. The structure of the characteristical borosilicate, danbourite, has been originally interpreted in the SiO_4-tetrahedra and the analogous BO_4-tetrahedra. A more detailed analysis showed that the B^{3+} ion should be regarded as having been shifted to one of the faces of the tetrahedron, which enables us to speak of a threefold coordination around the B^{3+} ion.

The structure of the boric acid $B(OH)_3$ itself is much more descriptive if it be represented as constructed of $B(OH)_4$ tetrahedra, which are then split into triangles and apexes entering into other triangles thus forming characteristical scales of boric acid.

It should be noted here that the typical trigonal CO_3-groups of calcite and aragonite have been shown to possess not a flat but a pyramidal (umbrella-shaped) structure, and it is only the incessant flapping of such an umbrella that makes it possible to speak of a flat CO_3-group, as an average of two pyramidal attitudes.

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STRATIGRAPHY

ON THE AGE OF THE SAKARAUL HORIZON

By I. A. KOROBKOV

(Communicated by A. A. Borissiak, Member of the Academy, 3. XII. 1938)

The name Sakaraul horizon was introduced in 1930 by Davitashvili(2) to designate, on ground of faunistic evidence, beds of sandstones, developed in Eastern Georgia, interstratified with clays of the Maikop series and rocks of Oncophora (Kotzakhur) beds.

The molluscan fauna of the Sakaraul horizon, represented by individuals of stenohaline species, among which large forms are of frequent occurrence, has not been known until very recent time. In literature one finds simply references to representatives of some or other genus present in the Sakaraul horizon. And only the work of Kharatashvili(4) on the geology of the Kura valley shows illustrations of shells of 6 species, of which 3 are determined as follows: *Turritella imbricataria*, *Pectunculus* sp. (cf. *P. obovatus* L.), *Cardium* sp. (cf. *Kubeckii* Hoern.), the rest being designated as: *Fusus* sp., *Melanopsis* sp., *Cardium* sp. It was certainly impossible, on the basis of inadequate data, to establish directly the age of the Sakaraul horizon, especially since the stratigraphic position of the latter might suggest both the Upper Oligocene and Lower Miocene periods.

In the summer of 1938, upon request of the Geological Oil Prospecting Institute, I have undertaken investigations of the stratigraphy, as well as colletions of fauna of mollusca from the Oligocene of Georgia. Simultaneousty the Sakaraul horizon has been investigated.

I have studied the Sakaraul horizon in outcroppings on the right and left bank of Kura. On the left bank rocks of that horizon are traceable throughout the region extending from the Gori mountain almost to the village Kaspi. On the right bank they stretch to the west of the quarries of the cement factory.

Over there, along the bank escarpment and in small intersecting ravines, the following section is observable. Brownish-grey shaley clays with fish scales and jarosite, with no visible signs of unconformity, are overlain by grey fine-grained consolidated sands, assuming a brownish tint by weathering, which contain shells of mollusks, mainly *Glycymeris*, *Meretrix*, *Pholadomya* and *Pinna*.

The sands display rounded concretions, isolated and linked like chains, of grey solid, often firmly compacted sandstone. The size of the concretions varies from 0.10—0.20 m to 5—6 m in diameter. There is either complete absence of fauna in the concretions, or they are crowded with fauna. Higher up the character of the concretions changes. The sandstone composing the concretions becomes less compacted, the molluscan fauna more abundant.

In one of the small ravines above the arenaceous beds, attaining 35 m in thickness, is deposited a pack of clays of Maikop habit, carrying siderite concretions, gypsum crystal druses and jarosite. Superimposed upon these clays, the visible thickness of which does not exceed 10—15 m, lie arenaceous sands identical with those recognized beneath the clays. The fauna of mollusks is less abundant in the concretions, while minute forms are of more frequent occurrence, as, for instance, *Corbula*, *Pectunculina*, *Cardium*. These rocks are replaced by a bed-pack of totally unfossiliferous sands 5 m in thickness, overlain by solidified sand with concretions, containing abundant vegetative remains. The rocks of the series of sands and sandstones resting above are utterly devoid of stenohaline species. Beginning from sand with vegetative remains, the rocks are gradually getting enriched with shells of *Oncophora*, *Eoprosodacna*, *Melanopsis*. The foregoing arenaceous beds are covered by the transgressively overlying Middle Sarmatian. From the foregoing section it may be seen that the Sakaraul horizon is bounded at the bottom by the clays of the Maikop series, and at the top by sandstones with vegetative remains, which, in my estimation, mark the beginning of Oncophora beds. Identical deposits and nearly the same lithological composition of the Sakaraul horizon may be seen also on the left bank of Kura.

The fauna of mollusks contained in the rocks of the Sakaraul horizon has a peculiar range of distribution. Thus, the lower sands display a prevalence of shells of *Glycymeris*, *Meretrix*, *Pholadomya* and *Pinna*. In the concretions the fauna is mostly distributed in such a way, that some concretions are crowded exclusively with shells of *Pectunculus*, other—with *Turritella*, some again—with *Cardium* and so forth. But on rare occasions shells referring to several genera were encountered in one concretion. Generally speaking, the molluscan fauna of the Sakaraul horizon is distinguished by paucity of generic and specific composition, though at the same time isolated specimens occur in prodigious numbers.

While studying the fauna of mollusks of the Sakaraul horizon, I have differentiated 15 species, of which 11 refer to the class *Lamellibranchiata* and only 4 species—to the *Gastropoda*, as enumerated below:

Meretrix (*Callista*) *erycinoides* Lamarck sp., *Meretrix* (*Cordiopsis*) *gigas* Lamarck sp., *Tapes* (*Callistotapes*) *vetulus* Basterot sp., *Tellina* (*Peronaea*) *planata* Linné, *Tellina* (*Moerella*) *donacina* Linné, *Corbula* (*Agina*) *gibba* Olivi sp., *Glycymeris menardi* Deshayes sp., *Phacoides* (*Linga*) *columbella* Lamarck sp. var. *basteroti* Agassiz sp., *Cardium echinatum* Linné, *Pectunculus* (*Axinaea*) *deshayesi* Mayer, *Limopsis* (*Pectunculina*) *anomala* Eichwald, *Fusus bilineatus* Partsch., *Pleurotoma duchastelli* Nyst, *Turritella* (*Haustator*) *turris* Basterot sp., *Pirula condita* Brongniart.

One new species has been established—*Discors* (?) *cartalinicum* n. sp.

The following have not been determined as species: *Tracia* sp., *Pholadomya* sp. (cf. *alpina*) *Chione* sp., *Cardium* sp. (n. sp. ?), *Pinna* sp., *Natica* sp.

The above-mentioned species show the following stratigraphic and geographic range of distribution (table).

The table illustrates that the majority of species representing the Sakaraul horizon have a wide range of stratigraphic distribution as well as of habitat; that 2 species had originated in the Upper Oligocene, 8 species—in the Aquitanian stage and 5 species in the Burdigalian. 14 species pass into the Helvetian stage, and only 8 into the Tortonian. It follows therefrom that the Sakaraul horizon, which carries a fauna most widespread in the Burdigalian and Helvetian stages, may be relegated to none but

Name of species	Name of stage				
	Upper Oligocene	Lower Miocene		Middle Miocene	
	Chattian	Aquitanian	Burdigalian	Helvetian	Tortonian
Meretrix (*Callista*) *erycinoides* Lamk. sp.	—	+	+	+	—
Meretrix (*Cordiopsis*) *gigas* Lamk. sp.	—	—	+	+	—
Tapes (*Callistotapes*) *vetulus* Bast. sp.	—	+	+	+	+
Tellina (*Peronaea*) *planata* Linné	—	+	+	+	+
Tellina (*Moerella*) *donacina* Linné	—	+	+	+	+
Corbula (*Agina*) *gibba* Olivi sp.	—	+	+	+	+
Glycymeris menardi Deshayes sp.	—	—	+	+	—
Phacoides (*Linga*) *columbella* Lamk. sp. var. *basteroti* Agassiz sp.	+	+	+	+	+
Cardium echinatum Linné	—	—	+	+	+
Pectunculus (*Axinaea*) *deshayesi* Mayer	—	+	+	+	—
Limopsis (*Pectunculina*) *anomala* Eichw.	—	—	+	+	+
Fusus bilineatus Partsch.	—	?	+	+	—
Pleurotoma duchastelli Nyst.	+	+	—	—	—
Turritella (*Haustator*) *turris* Basterot sp.	—	+	+	+	+
Pirula condita Brongniart	—	+	+	+	—
	2	10 (11?)	14	14	8

these stages. Furthermore, if one considers that the Sakaraul fauna includes a species (*Pleurotoma duchastelli* Nyst) widely spread in the Middle and Upper Oligocene and not reaching above the Aquitanian stage, evidence is gained in favour of the Burdigalian stage. The evidence becomes still more conclusive upon a quantitative analysis of occurrence of individual representatives of species. Thus, in the Sakaraul horizon are more frequently found shells of *Meretrix* (*Cordiopsis*) *gigas* Lamk., *Glycymeris menardi* Deshayes, *Pectunculus* (*Axinaea*) *deshayesi* Mayer, *Turritella* (*Haustator*) *turris* Bast. In Western Europe the former 3 species occur most frequently in the Burdigalian stage, and the latter species is distributed equally in the Burdigalian stage and the Helvetian. Consequently, the described analyses of Sakaraul mollusks lead to the suggestion that the Burdigalian is the most probable age of the Sakaraul horizon.

At the present time I am about to complete a study of another complex of molluscan species of the Lower Miocene, discovered by the geologist E. K. Vakhanya in Abkhazia in the clays of the upper horizons of the Maikop series exposed in the quarries of the brick factory at the village Quezani. This complex comprises 23 species, which on the basis of a stratigraphic analysis permit to assign the strata enclosing the aforesaid fauna to the Burdigalian stage. The complex concerned includes 10 species, revealed in the Sakaraul horizon.

These 10 species serve as guide-species for identification of rocks of the deposits under comparison. Thus, the Sakaraul horizon and the strata con-

taining *Pectunculus* of Abkhazia may be considered as coeval formations. The latter circumstance offers additional support to the assigning of the Sakaraul horizon to the Burdigalian stage.

Likewise, the stratigraphic position of the studied horizon contributes toward the identification of its age. The rocks of the Sakaraul horizon, with no signs of unconformity, are overlain by the arenaceous complex of Oncophora or Katzakhur beds. Without launching into a broad literary survey on the question of the age of Oncophora beds*, it should be noted that the majority of investigators concur in the opinion regarding the parallelism of the Oncophora beds with the Grundian horizon, i. e. relegating the former to the Helvetian stage.

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* Details on the stratigraphy and fauna of the Katzakhur horizon are contained in works by Davitashvili ([1], [2], [3]).

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